

## LENS ARRAY SHEET

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FIELD OF THE INVENTION

The present invention relates to a lens array sheet suitable to a light condensing plate used for a display device of an organic electroluminescence element and liquid crystal, etc., a mold suitable for being used for producing the lens array sheet, a light condensing plate composed of the lens array sheet, an organic electroluminescence illuminant comprising the lens array sheet, and a display device comprising the illuminant.

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BACKGROUND ART

There are demands for a surface lighting device used for a display device to have many characteristics, such as high luminance, low power consumption and thin shape. When improving one of these characteristics, it is liable that other performance has to be more or less sacrificed and it has been very difficult to improve all performances at a time.

To overcome the plurality of challenges, a sheet (lens array sheet) having a lens array formed with a fine concave and convex pattern, such as a prism shape and a

Fresnel lens shape, on a surface of a thermoplastic resin mold, such as a transparent plastic, has been increasingly used as an optical part in recent years.

For example, a method of easily attaining high  
5 luminance by using a lens array sheet as a light  
condensing plate for a liquid crystal display device and  
condensing a display light in an approximately normal  
direction has become widely used. This method uses a  
light condensing effect of a prism and is capable of  
10 attaining high luminance without increasing an electric  
power and a thickness of the device, while it has a  
disadvantage that the luminance drastically declines when  
being out of the light condensing range.

To solve such a disadvantage, the Japanese  
15 Unexamined Patent Publication No. 7-261006 proposes to  
change a refraction index between one surface and the  
other surface of the prism. Also, the Japanese Unexamined  
Patent Publication No. 2000-75102 proposes to use as a  
material of the light condensing plate an alicyclic  
20 structure containing thermoplastic resin having a  
norbornene structure.

Since an organic electroluminescence material can  
be used as a thin film, it is an effective material for  
making a surface lighting device and a display device  
25 thinner and lighter. However, light transmittance of a

liquid crystal panel is too low to use it as a backlight of a liquid crystal display device, therefore, the light condensing efficiency has to be furthermore improved comparing with that in a conventional backlight using a cold-cathode tube, etc.

However, when using a prism lens having a line shaped surface, which has been conventionally used as a light condensing plate of a liquid crystal substrate, as a light condensing plate of an organic electroluminescence element, there is a disadvantage that the viewing angle becomes narrow.

#### SUMMARY OF THE INVENTION

An object of the present invention is to provide a lens array sheet having a wide viewing angle and an improved light condensing efficiency, a mold suitable for being used for producing the lens array sheet, a light condensing plate comprising the lens array sheet, an organic electroluminescence element comprising the lens array sheet, and a display device comprising the element.

The present inventors confirmed that a line shaped prism lens converges a light only in one direction and that the disadvantages explained above arise. As a result that they have been committed themselves to study for attaining an improvement in light convergence, they found

that a lens array sheet having pyramid-shaped projections and recesses is capable of most effectively condensing even a light from an organic electroluminescence element comparing with ones having projections and recesses in a three-sided pyramid, six-sided pyramid, eight-sided pyramid or conical shape, so that the present invention has completed.

According to the present invention, there is provided a lens array sheet having a plurality of pyramid-shaped projections or recesses on a surface of its transparent base material film.

Preferably, a bottom surface of the pyramid shape is a rectangular or square shape satisfying a relationship of (length "a" of one side)  $\leq$  (length "b" of other side)  $\leq 10a$ .

Preferably, the length "a" of one side of the bottom surface of the pyramid shape is 0.1  $\mu\text{m}$  to 20  $\mu\text{m}$ .

Preferably, a height "c" of the pyramid shape is  $0.2a \leq c \leq 2a$  with respect to the length "a".

Preferably, a width "s" between adjacent projections or recesses is not more than 50% of the length "a".

Preferably, a base angle  $\theta$  of side surfaces of the pyramid shape is  $20^\circ$  to  $80^\circ$ .

Preferably, the transparent base material film is

composed substantially of an alicyclic olefin resin. The expression "composed substantially of a transparent resin material" means the case of composing all by a transparent resin material and the case of including various additives in accordance with need as far as it is not more than 50 wt% or so with respect to the total amount.

Preferably, the alicyclic olefin resin is a norbornene based polymer or a vinyl alicyclic hydrocarbon polymer.

Preferably, the lens array sheet is produced by injection molding using a mold having pyramid-shaped projections or recesses on its surface.

According to a first aspect of the present invention, there is provided a mold, made by a metal layer and provided with pyramid shaped projections on its surface, wherein said metal layer is obtained by forming a metal layer on said pattern of a substrate formed with a pyramid-shaped concave pattern and peeling the metal layer from the substrate. In the first aspect, it is preferable to use as the substrate formed with a pyramid-shaped concave pattern a mold made by silicon formed by (1) a step of forming a positive type resist pattern on a silicon wafer formed on its surface with an silicon oxide layer, (2) a step of forming a silicon oxide pattern by

performing etching on the silicon oxide layer by an etching solution containing hydrofluoric acid by using the resist pattern as a mask, (3) a step of removing the resist pattern and performing anisotropic etching on the silicon wafer surface by an alkaline solution to form  
5 pyramid-shaped recesses, and (4) a step of removing the silicon oxide pattern by an etching solution containing hydrofluoric acid.

According to a second aspect of the present invention, there is provided a mold made by a metal layer and provided with pyramid shaped projections on its surface, wherein

said metal layer is obtained, by using the mold as set forth in claim 10 or 11, by forming a metal layer on  
15 a surface of the mold and peeling the metal layer from the mold.

According to the present invention, there is provided a light condensing plate composed of the above lens array sheet.

20 According to the present invention, there is provided an organic electroluminescence element, comprising a transparent substrate composed of the above lens array sheet, a transparent electrode layer stacked on the transparent substrate, an organic  
25 electroluminescence material layer stacked on the

transparent electrode layer, and a metal electrode layer stacked on the organic electroluminescence material layer.

According to the present invention, there is provided a display device comprising the above organic  
5 electroluminescence element.

#### BRIEF DESCRIPTION OF DRAWINGS

FIG. 1A, FIG. 2A, FIG. 3A and FIG. 4A are plan views of examples of a pyramid shape;

10 FIG. 1B, FIG. 2B, FIG. 3B and FIG. 4B are views from the front of the respective examples in FIG. 1A, FIG. 2A, FIG. 3A and FIG. 4A;

FIG. 5A, FIG. 5B and FIG. 5C are plan views showing examples of pyramid-shaped projections or recesses formed  
15 on a transparent base material film;

FIG. 6A and FIG. 6C are sectional views of examples of pyramid-shaped projections or recesses formed on a transparent base material film, wherein the projections or recesses are formed to be in a regular lattice shape;  
20 and

FIG. 6B and FIG. 6D are sectional views of examples of pyramid-shaped projections or recesses formed on a transparent base material film, wherein the projections or recesses are formed to be in a hound's-tooth check  
25 shape (note that a dotted line indicates pyramid-shaped

projections or recesses on the next line).

Note that as to references in the drawings, "a" indicates a length of a side forming a bottom surface of a pyramid shape, "b" indicates a length of other one side forming the bottom surface of the pyramid shape, "c" indicates a height of pyramid-shaped projections or recesses,  $\theta$  indicates a base angle of side surfaces of the pyramid shape, 1 indicates pyramid-shaped projection or recess, 2 indicates a lens array sheet, and "s" indicates a width between pyramid shaped projections or recesses.

#### DESCRIPTION OF THE PREFERRED EMBODIMENT

Below, the present invention will be explained based on embodiments shown in drawings.

##### Lens Array Sheet

A lens array sheet of the present invention has pyramid-shaped projections or recesses on a surface (preferably, on one surface) of a transparent base material film.

##### Transparent Base Material Film

A transparent base material film to be used in the present invention is a film shaped mold composed substantially of a transparent resin material. The expression "composed substantially of a transparent resin



material" means the case of composing all by a transparent resin material and the case of including various additives in accordance with need as far as it is not more than 50 wt% or so with respect to the total amount.

As specific examples of the transparent resin material, polyethylene, polypropylene, polymethylpentene and other linear polyolefin resins; polystyrene and other aromatic vinyl based resins; a norbornene based polymer, a vinyl alicyclic hydrocarbon polymer, a monocyclic olefin based polymer, a cyclic diolefin based polymer and other alicyclic olefin resins; polycarbonate, polyethylene phthalate, polybutylene phthalate, liquid crystal polyester and other polyester based resins; polymethyl methacrylate and other acrylic resins; an acrylonitrile styrene resin, an acrylonitrile styrene butadiene resin and other acrylonitrile based resins; polysulfone, polyether sulfone, polyphenylene sulfide, polyphenylene ether and other polyether based resins; and polyether ether ketone and other ketone based resins; may be mentioned.

Among them, a linear polyolefin resin, an alicyclic olefin resin and polyether based resin, etc. are preferable because they easily bring cross-linking reaction by irradiation of an active energy beam and

solubility in specific solvents easily changes.

Furthermore, an alicyclic olefin resin is the most preferable because of less absorption of visible light and a low water-absorbing property.

5           As specific examples of an alicyclic olefin resin, a ring-opened polymer of a norbornene based monomer and the hydrogenated product, a ring-opened polymer of other monomer capable of copolymerizing with a norbornene based monomer and the hydrogenated product, an addition polymer  
10 of a norbornene based monomer and the hydrogenated product, an additional copolymer with other monomer capable of copolymerizing with a norbornene based monomer and the hydrogenated product and other norbornene based polymers; polyvinylcycloalkane, polyvinylcycloalkene,  
15 hydrogenated product of an aromatic vinyl polymer and other vinyl alicyclic hydrocarbon polymer; polycyclopentene, polycyclohexene and other monocyclic olefin based polymers; polycyclohexadiene and other cyclic diolefin based polymers; etc. may be mentioned.  
20 Among them, a norbornene based polymer and a vinyl alicyclic hydrocarbon polymer are the most preferable. Also, a vinyl alicyclic hydrocarbon polymer may be a copolymer with other monomer (for example, butadiene, isoprene and other vinyl based monomers, etc.) capable of  
25 copolymerizing with vinylcycloalkane, vinylcycloalkene

and an aromatic vinyl based monomer, etc., and the polymer form may be either of a block polymer and a random polymer.

A thickness of the transparent base material film  
5 is normally 0.05 to 3 mm or so.

#### Pyramid-shaped Projections or Recesses

Projections or recesses on a surface of the transparent base material are pyramid shaped.

The projections or recesses may have a pyramid  
10 shape having different base angles of side surfaces or a right pyramid shape wherein all of the bottom angles are the same. Alternately, it may be a pyramid shape wherein an apex angle is cut off. Specifically, shapes shown in FIG. 1A, FIG. 1B, FIG. 2A, FIG. 2B, FIG. 3A, FIG. 3B, FIG.  
15 4A and FIG. 4B are the examples but it is not limited to these.

Also, in terms of improving front luminance by improving the light condensing property, it is preferable that the bottom surface of the pyramid shape is a  
20 rectangular or square shape satisfying the relationship below. The relationship is preferably (length "a" of one side)  $\leq$  (length "b" of other side)  $\leq 10a$ , more preferably (length "a" of one side)  $\leq$  (length "b" of other side)  $\leq 5a$ , further preferably (length "a" of one side)  $\leq$  (length  
25 "b" of other side)  $\leq 2a$ , particularly preferably (length

"a" of one side)  $\leq$  (length "b" of other side)  $\leq 1.5a$ .

The length "a" of one side of the bottom surface of the pyramid shape is not particularly limited but is normally 0.1  $\mu\text{m}$  to 500  $\mu\text{m}$ , preferably 0.1  $\mu\text{m}$  to 100  $\mu\text{m}$ .

5 Particularly in the case of obtaining an organic electroluminescence element by using a lens array sheet of the present invention as a light condensing plate and using it as a backlight of a liquid crystal display device, the length "a" is preferably 0.1  $\mu\text{m}$  to 20  $\mu\text{m}$ , so  
10 that the liquid crystal is capable of preventing interference by an outside light (so-called moiré).

The base angle  $\theta$  of side surfaces of the pyramid shape is preferably  $20^\circ$  to  $80^\circ$ , particularly preferably  $30^\circ$  to  $75^\circ$ .

15 A height "c" of the pyramid shape (a length from the bottom surface to the apex angle) is not particularly limited and is preferably  $0.2a \leq c \leq 2a$ , more preferably  $0.5a \leq c \leq 1.5a$  with respect to the length "a" of one side of the bottom surface of the pyramid shape.

20 It is sufficient when a large number of the projections and recesses as above are arranged on a surface of a transparent base material film, and directions of the respective projections and recesses may be a regular lattice shape as shown in FIG. 5A, a  
25 hound's-tooth check shape as shown in FIG. 5B, or a

random shape as shown in FIG. 5C. A regular lattice shape or hound's-tooth check shape is preferable for obtaining higher luminance.

Also, a shape of the respective projections or  
5 recesses arranged on the surface of the transparent base material film may be all same or independent shapes from one another.

Production Method of Lens Array Sheet (Formation  
Method of Projections or Recesses)

10 A method of forming pyramid-shaped projections or recesses on a surface of the transparent base material film is not particularly limited. As typical techniques, for example, (1) a method of setting a mold formed with pyramid-shaped projections or recesses to perform  
15 injection molding, compression molding or heat-melt molding, such as blow molding, (2) a method of using a die formed with pyramid-shaped projections or recesses to perform melt extrusion molding, (3) a so-called 2P method for applying an ultraviolet ray curing resin on a molding  
20 surface formed with pyramid-shaped projections or recesses, then, curing the resin by irradiating an ultraviolet ray to transfer a pattern, may be mentioned. Among them, a method of transferring a pattern by using the above method (1) is preferable.

25 A method of producing a mold is not particularly

limited. For example, to produce a mold having pyramid-shaped "projections" on its surface, a method of forming a metal layer by stacking a metal by a soldering method or a sputtering method, etc. on a substrate formed with a pyramid-shaped concave pattern, then, peeling the metal layer from the substrate may be mentioned.

To obtain a mold having pyramid-shaped "recesses" on its surface, a method of using a mold having pyramid-shaped projections obtained as above and performing mold release processing for bringing the mold surface into contact with potassium bichromate or other oxidizing agent, etc. in accordance with need, then, forming a metal layer by the same method as above by stacking metal, and peeling the metal layer from the mold may be mentioned.

A method of obtaining a substrate formed with a pyramid-shaped concave pattern is not particularly limited, and a method of using a property that a silicon single crystal substrate is anisotropically etched by an alkaline solution (etching solution) may be mentioned as a preferable example because a fine pattern is easily obtained. Specifically, this method is by (1) stacking an oxidized silicon film and a photo resist film on a silicon substrate in this order, then, forming a pattern on the photoresist film, (2) performing etching on the

oxidized silicon layer by an etching solution containing hydrofluoric acid by using the resist pattern as a mask to form a silicon oxide pattern, (3) furthermore, performing anisotropic etching on the silicon substrate by a sodium hydroxide solution or other alkaline etching solution to form pyramid-shaped recesses on the silicon substrate (at this time, the resist pattern can be also removed at a time for it is soluble in the alkaline etching solution), and (4) bringing the finally left silicon oxide pattern into contact with an etching solution containing hydrofluoric acid to remove the silicon oxide pattern, projection or recesses (pattern) is formed on the surface of the silicon substrate.

When forming pyramid-shaped projections or recesses on a surface of a transparent base material film by using a mold produced by the above explained method, a width of a resist pattern to be a mask becomes a width "s" between adjacent projections or recesses on a lens array sheet of the present invention. The narrower the width "s" is, the more preferable. It is preferably 50% or less, more preferably 20% or less, further preferably 10% or less and particularly preferably 5% or less with respect to a length "a" of one side of the bottom surface of the pyramid shape.

### Electroluminescence Element

A light condensing plate of the present invention is made by the above lens array sheet.

An organic electroluminescence element of the present invention is obtained by stacking a transparent electrode layer, such as indium tin oxide (ITO), an organic electroluminescence material layer including an organic element, and a metal electrode layer in this order on the lens array sheet as a transparent substrate. In the organic electroluminescence element, the lens array sheet functions as a light condensing plate.

The organic electroluminescence element can be used as a backlight of a liquid crystal display device, etc.

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### EXAMPLES

Next, specific examples of the embodiment of the present invention will be taken to explain the present invention more in detail. Note that the present invention is not limited to the examples.

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#### Example 1

##### Production of Substrate

After applying a positive type photoresist composition (product name: ZPP1700PG) made by ZEON Corporation by spin-coating on a substrate obtained by forming a film of SiO<sub>2</sub> to 300Å on a silicon, the result

25



was prebaked at 100°C to obtain a resist film of 1.5  $\mu\text{m}$  on the substrate.

The obtained resist film was subjected to exposure of 50 mJ/cm<sup>2</sup> by an exposure apparatus "PLA501F" made by  
5 Canon Inc. via a mask, development processing was performed by a 2.38% tetramethyl ammonium hydroxide solution for 60 seconds, and then, rinse processing by ultrapure water was performed for 30 seconds. The substrate was dried by spin processing and prebaking  
10 processing at 120°C was finally performed to obtain a resist pattern on the substrate.

The thus obtained substrate was dipped in a hydrofluoric acid buffer solution (mixture of 3.6% hydrofluoric hydrofluoric water and 18% hydrofluoric  
15 ammonium water by 1:1 (in volume) at 20°C. It will be the same below.) for five minutes to etch SiO<sub>2</sub>, rinsing processing for immersing the etched substrate in pure water for 60 seconds was performed, then, the substrate was dried by spin processing.

20 The substrate, wherein the oxide silicon film was etched, was immersed in a 30% sodium hydroxide solution for 30 minutes at 80°, immersed in pure water for 60 seconds, then, dried by blowing a dry air, so that pyramid-shaped concave recesses were formed on the  
25 silicon substrate.

To remove an excessive oxide silicon film remaining on the silicon substrate formed with the recesses, the substrate was dipped in a hydrofluoric acid buffer solution for 5 minutes, then, immersed in pure water for 5 60 seconds, and dried by blowing a dry air, so that a silicon substrate having pyramid-shaped recesses was produced.

#### Production of Mold

After adhering the obtained silicon substrate on a 10 fixture and forming a nickel film of about 500Å by vacuum evaporation on the surface, nickel metal was grown by performing electrolytic soldering in an electrolytic solution containing nickel sulfamate as its main component. The obtained nickel block (metal layer) was 15 peeled from the silicon substrate, so that a mold (convex mold) formed with pyramid-shaped "projections" in a hound's-tooth check shape was produced.

#### Production of Lens Array Sheet

This mold was set in an injection molding apparatus 20 to obtain a lens array sheet (the outside dimension is 40 mm × 40 mm) having a thickness of about 1 mm by injection molding (resin melt temperature of 285°C, mold temperature of 130°C, and resin filling time of about 0.2 second) using a cycloolefin polymer (ZNR1430R made by 25 ZEON Corporation).

The obtained lens array sheet was formed on its surface with pyramid-shaped "recesses" having a bottom surface of  $20\text{ }\mu\text{m} \times 20\text{ }\mu\text{m}$ , a base angle of side surfaces of approximately  $55^\circ$  and a height of  $14\text{ }\mu\text{m}$  in a hound's-tooth check shape.

#### Production of Organic Electroluminescence Element

ITO having a thickness of  $300\text{\AA}$  was evaporated on the thus obtained lens array sheet, the result was washed with steam of IPA, set in an organic electroluminescence element producing apparatus, and subjected to plasma processing at 100W under an atmosphere of oxygen/argon = 50/50 for 5 minutes.

After that, the result was transferred to an organic evaporation chamber, and N,N-di(1-naphthyl)-N,N'-diphenyl-1,1'-diphenyl-1,4-imine (NPD) as a hole transport material and 8-hydroxy quinoline aluminum (Alq3) as an electron transport material were put in two melting pots wound by coil. Then, inside the chamber was depressurized to  $10^{-6}\text{ Pa}$  and a current of 20A was applied to the coil holding NPD to perform evaporation until the film thickness becomes  $400\text{\AA}$ , then, evaporation was performed under the same condition until a film thickness of Alq3 becomes  $600\text{\AA}$ , so that an organic electroluminescence material layer was stacked.

After that, the substrate was moved to a metal

evaporation chamber while keeping in the system in a vacuum state, and lithium fluoride loaded on a metal board was heated, so that the lithium fluoride was evaporated to be 5Å on the organic layer. Then, aluminum  
5 was loaded on another metal board to evaporate it to be a thickness of 1000Å by the same operation, so that a stacked body, wherein an ITO layer, an organic electroluminescence material layer and a metal electrode layer were stacked, was obtained on the lens array sheet.

10 The thus obtained stacked body was attached with a stainless sealing tube applied with an ultraviolet ray curing adhesion in a globe box in a dry nitrogen, and an ultraviolet ray was irradiated to adhere the sealing tube, consequently, an organic electroluminescence element was  
15 obtained.

#### Evaluation of Element

Measurement of luminance was made on the obtained element by a luminance measurement device (BM-8 made by TOPCON CORPORATION) in a state of being placed vertical  
20 with respect to the substrate surface, and the result was 2165 cd/cm<sup>2</sup>. When luminance was measured in the same way in a state of giving a tilt by 45° with respect to the substrate surface respectively to above, below, right and left (evaluation of a viewable angle), the result was  
25 above: 2100 cd/cm<sup>2</sup>, below: 2120 cd/cm<sup>2</sup>, left: 2135 cd/cm<sup>2</sup>

and right 2140 cd/cm<sup>2</sup>. Namely, sufficient luminance was obtained even when viewing from the directions tilted by 45° with respect to the substrate surface, so that it was confirmed that a sufficient viewable angle was attained.

5           Comparative Example 1

Other than using a film of a cycloolefin polymer (ZNR1430R made by ZEON Corporation) having a thickness of 1 mm and not formed with concaves and convexes instead of the lens array sheet of the example 1, an organic  
10 electroluminescence element was produced in the same method as in the example 1, and the luminance was measured under the same condition. The result was 1250 cd/cm<sup>2</sup>. Accordingly, it was confirmed that by using the lens array sheet of the example 1 as a light condensing  
15 plate, luminance of the organic electroluminescence element was improved to about 1.5 times.

Also, evaluation of the viewable angle measured in the same way was above: 1000 cd/cm<sup>2</sup>, below: 955 cd/cm<sup>2</sup>, left: 990 cd/cm<sup>2</sup> and right 930 cd/cm<sup>2</sup>.

20           Example 2

The mold in the example 1 was used. The mold was immersed in a potassium bichromate solution (0.1 wt%) for 30 seconds and mold releasing processing was performed by oxidizing the mold surface, then, a nickel layer (metal  
25 layer) was stacked under the same condition as that in

the example 1 and the stacked nickel layer was peeled from the mold, so that a mold (concave mold) formed with pyramid-shaped "recesses" having a base angle of side surfaces of approximately  $55^\circ$  and a height of  $14\text{ }\mu\text{m}$  in a hound's-tooth check shape was obtained.

By using the obtained mold, a lens array sheet was produced in the same way as in the example 1. The obtained lens array sheet was formed on its surface with pyramid-shaped "projections" having a bottom surface of  $20\text{ }\mu\text{m} \times 20\text{ }\mu\text{m}$ , a base angle of side surfaces of approximately  $55^\circ$  and a height of  $14\text{ }\mu\text{m}$  in a hound's-tooth check shape.

By using the lens array sheet, an organic electroluminescence element was produced, and the luminance was measured under the same condition. The result was  $1420\text{ cd/cm}^2$ . From the result, it was confirmed that luminance of the organic electroluminescence element was improved to about 1.7 times comparing with that in the comparative example 1 by using the lens array sheet of the example 2 as a light condensing plate.

Also, evaluation of the viewable angle measured in the same way was above:  $1400\text{ cd/cm}^2$ , below:  $1395\text{ cd/cm}^2$ , left:  $1405\text{ cd/cm}^2$  and right  $1380\text{ cd/cm}^2$ , and superiority of the example 2 to the comparative example 1 was confirmed.

### Analysis

The followings are understood from the examples 1, 2 and the comparative example 1 above. When using as a light condensing plate a lens array sheet formed with a plurality of projections or recesses on one surface of a transparent base material film, luminance of an organic electroluminescence element improves. Particularly, by choosing "recesses" from the projections and recesses to form, higher luminance is obtained.

10        A viewing angle was confirmed to have a tendency of becoming wider as the luminance improves.

### INDUSTRIAL APPLICABILITY

A lens array sheet of the present invention exhibits remarkable efficiency in improving luminance as a light condensing plate of an organic electroluminescence element.

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The lens array sheet of the present invention can be used as an optical part, such as an information recording medium, optical lens, optical filter, light guide plate for a liquid crystal display device and optical sheet, in addition to being used as a light condensing plate of an organic electroluminescence element.

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